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DEVELOPMENT AND APPLICATION OF CHARCOAL SORBENTS FOR CRYOPUMPING FUSION DEVICES

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Progress has been made in defining the capabilities of charcoal as the most promising absorbent to be used in cryopumps for fusion power application. The capabilities of alternative methods of cryopumping helium have been examined in a literature survey and by test, and the results are described here. Considerations include pumping speed, capacity to accumulate pumped gas, ease of reconditioning, use of alternative materials and tolerance to the fusion environment. Vacuum pumps for future fusion devices must handle large quantities of helium/hydrogen isotopes and other impurities. Cryopumps or turbomolecular pumps have demonstrated the capability on a small scale, and each has an important advantage: TMPs do not accumulate gases; cryopumps can separate helium from other effluents.

This paper includes a review of a method for selecting charcoals for helium cryopumping, testing of a continuously operating cryopump system, and definition of a design that is based on the requirements of the Next European Torus. Tritium limits are satisfied. The pump design incorporates the charcoal sorbent system that has been recently developed and is based on a reasonable extrapolation of current state-of-the-art. Evaluation of alternative methods of separating helium and other gases led to selection of a movable barrier as the preferred solution.

1. Introduction

A program was undertaken to develop high performance, compatible charcoal sorbent systems for pumping helium and other constituents exhausted from fusion experiments and reactors. The program described in ref. [1] included the following key elements:

- A survey by test of charcoals resulted in the selection of coconut charcoal as best for the application.
- A survey by test of charcoal sizes resulted in the selection of an intermediate size. Of those surveyed, 0.6–1.7 mm screen size produced the highest performance.
- An evaluation of different methods of attaching charcoal to metal substrates resulted in the selection of a copper cement and a silver–copper–phosphorus braze which provided adhesion without adversely affecting the charcoal's sorption capability.
- A demonstration showed that charcoal could be regenerated after prolonged exposure and contamination at ambient conditions in a passive vacuum. Specification of a procedure to avoid such contamination was produced.
- The AgCuP braze/coconut charcoal was applied to a 40.6 cm diameter liquid helium dewar for installation in a compound cryopump to be tested at the Tritium Systems Test Assembly (TSTA) at Los Alamos National Laboratory in 1988.

This paper describes the continued effort to develop, demonstrate, and characterize the charcoal system for fusion application.

2. Characterization of charcoals

The value of charcoal as a sorbent derives from its extensive network of microscopic pores, and its wide distribution of pore sizes. These characteristics vary widely between charcoal types, e.g., coconut, coal, wood. Significant variations also exist within a single type. The capability to cryopump helium was determined for a selection of charcoals, tabulated in table 1. Samples were bonded with a silver epoxy to 10.2 cm diameter aluminum plates. These were, in turn, attached to the surface of a liquid helium dewar inside a test pump. The samples were separated by a chevron from the pressure gauge used for pumping speed calculations. The chevron was sufficiently large to maximize conductance when compared to the size of the sample.

Fig. 1 shows a comparison of pumping speeds for the selected charcoals. The charcoal shown in ref. [1] to

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Table 1

TYPE	SOURCE	SIZE U.S. MESH	SIZE SI UNITS (mm)	SUPPLIER	CALCULATED TOTAL SURFACE AREA (m^2/g)	FRACTION OF BET SURFACE AREA
PCB	COCONUT	12 X 30	1.7–0.6	CALGON (PITTSBURGH)	548	0.49
BPL	COAL	12 X 30	1.7–0.6	CALGON	723	0.70
965	COKE SLUDGE BY-PRODUCT	12 X 20	1.7–0.8	WITCO CHEMICAL (NEW YORK)	800	0.72
GAC 102 GA	COAL	10 X 25	2.0–0.7	CECA, INC. (TULSA)	1428	1.16
GAC 1240	COAL	12 X 40	1.7–0.4	CECA, INC.	862	0.85
NUCHAR WV-B	HARDWOOD	10 X 25	2.0–0.7	WESTVACO (COVINGTON, VA)	1880	1.20

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be the best for pumping helium, was again the best performer in this expanded evaluation. Interestingly, the charcoals having the highest pore volume and pore surface area based on a standard nitrogen sorption test, yielded poor helium pumping performance of the test samples. Clearly these characteristics are not appropriate indicators of the charcoal's value as a cryosorbent for helium.

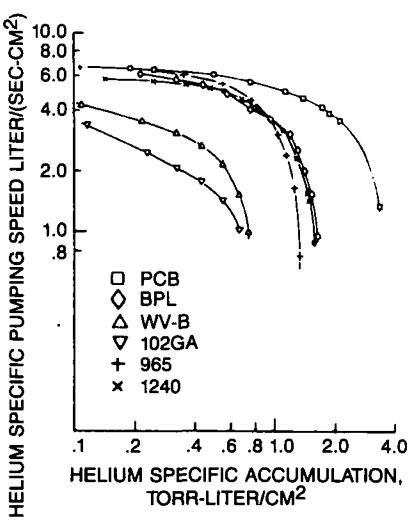

Fig. 1. Helium pumping performance for selected charcoals.

A further evaluation of the nitrogen sorption data showed, for some samples, a significant deviation between the value of the total pore area determined from the standard nitrogen sorption tests, and that derived from the BET method [2]. These areas should be equal; in practice, however, it is common to see a variation of ±20% between the two values. The nitrogen sorption test used in the subject program accounts for pore sizes greater than approximately 14 Å (1.4×10^{-7} cm); the BET method accounts for all pore sizes. Presumably any difference outside the expected variation can be the result of the presence of many pores in the range below 14 Å. These are the pore sizes most likely to trap helium.

Note that the best helium cryo-sorber (PCB coconut) shows a significant deviation between the areas derived by the two methods, indicating for it a large population of pore sizes below 14 Å. By contrast, the poorer cryo-sorber (WV-B hardwood) has an expected variation range, which is shown in a comparison of the areas determined by the two methods. This finding indicates that most of its pore sizes are in the range above 14 Å. Thus, of these two, the sample with the higher pore area was the poorer absorber of helium, and the sample with the larger count of small pore sizes was the better choice for pumping helium.

Fig. 2 compares nitrogen sorption isotherms for these two charcoals. Relative pressure, i.e., the test pressure ratioed to 1 atm (10^5 Pa), is indicative of pore size distribution. The coconut holds more gas when the

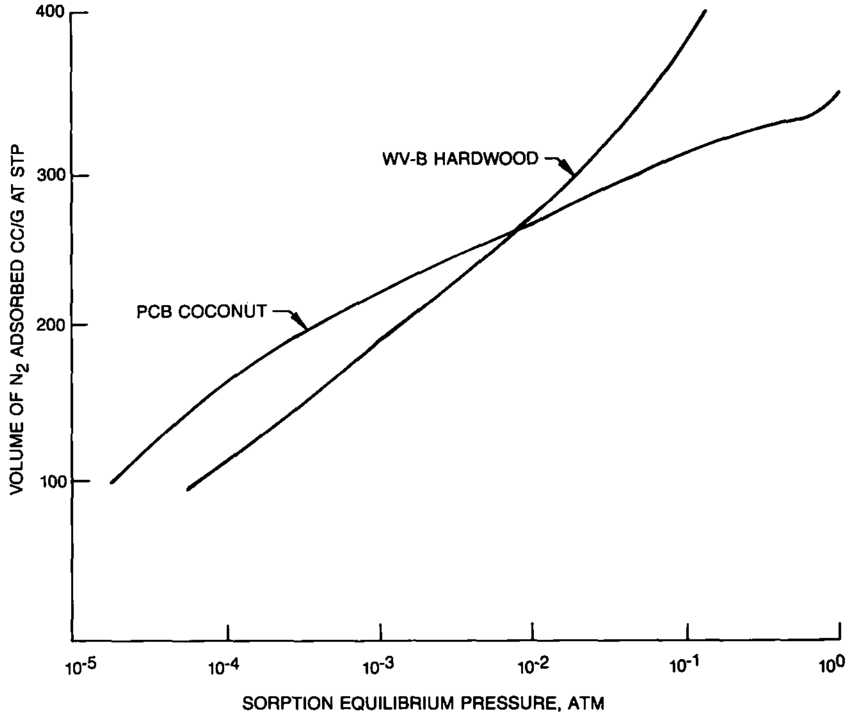

Fig. 2. N_2 isotherms for charcoals with extremes of helium cryopumping capability (per fig. 1).

smaller pores are filling, while the hardwood holds more gas overall. A charcoal suitable for helium cryopumping is, therefore, one that follows the sorption characteristics of the coconut charcoal.

A comparison of the charcoal's helium pumping performance and its ability to absorb nitrogen might indicate that a relatively low-cost nitrogen sorption test offers a method of screening candidate charcoals for use in helium pumps. In the absence of more definitive results, a charcoal that absorbs 100 or more standard cm^3/g at a test pressure of 1.7 Pa in a standard nitrogen sorption test is a useful sorbent for helium cryopumping.

3. Continuous cryopumping of helium

Ref. [1] described the development of the fusion-compatible charcoal bonding systems and included a description of a cryopump system designed for continuous, automatic pumping and regeneration of deuterium. The pumping system is described fully in ref. [3]. This condensation pump was subsequently modified to pump helium by the addition of charcoal to the liquid helium-cooled pumping panels (fig. 3).

Tests demonstrated the ability of the continuous duty cryopump (CDC) to continuously pump helium, from the time of the operator's start command at the computer keyboard until the test was ended by switching to manual mode. The helium sorption rate was constant during the automatic 35 min pumping periods of each section. Throughput was held at 1.7×10^{-5} Pa m^3/s (1.3×10^{-4} Torr l/s) per sq cm of charcoal

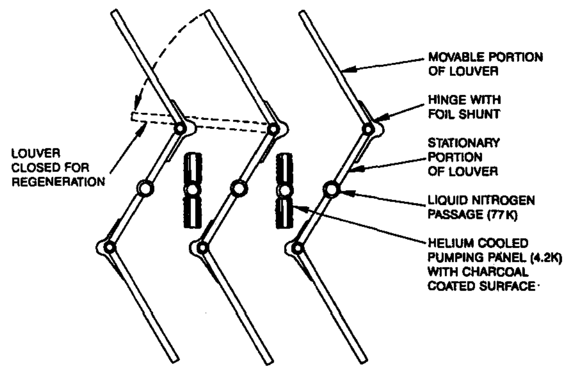

Fig. 3. Location of charcoal in the continuous duty chamber cryopump.

surface area. Vacuum chamber pressure, which was maintained at 2.3×10^{-3} Pa (1.7×10^{-5} Torr) during pumping, rose to 0.13 Pa (10^{-3} Torr) during regeneration, indicating that a portion of the gas being pumped by the roughing system actually returned to the chamber through the pump's closed louvers. This back-leakage was excessive, although previous tests with deuterium test gas resulted in satisfactory levels. Extensive effort is required to properly adjuste the pump's moving chevrons that form the closure during regeneration, and this was not undertaken. The back-leakage objective had been met previously, but the result showed the need to minimize in future designs the length of the sealing surface, which typically operates at cryogenic temperatures.

The CDC was operated over a range of constant helium throughputs from 1.7×10^{-5} to 2.0×10^{-6} Pa m^3/s (1.3×10^{-4} to 1.5×10^{-3} Torr l/s) per sq cm of charcoal to assess the effect of throughput on pumping speed. Data taken within two minutes of the start of each run to negate the effect of accumulation of helium on the charcoal showed that speed was practically unaffected. Helium pumping speed was 7.4 m^3/s compared to a deuterium pumping speed of 17.4 m^3/s measured before coating the pumping surfaces with charcoal. The difference is in part accounted for by the fact that only 44% of the total surface cooled to liquid helium temperature, which could pump deuterium, was coated with charcoal for pumping helium.

A series of runs was made in which helium and hydrogen were co-pumped on the charcoal and on the condensing surfaces of the pump. The hydrogen–helium mixture throughputs were held constant at 7.7 : 1 or 18 : 1 by volume, with the hydrogen flow at 1.33×10^{-4} Pa m^3/s (10^{-3} Torr l/s) per sq cm of charcoal. A preliminary test with hydrogen only yielded a pumping speed of 21.3 m^3/s. Pressure surges occurred at both mixture rate conditions, indicating spurious release from the pumping surface. The extent of the CDC mixture tests was insufficient to allow assessment of the cause of these releases. However, the need to resolve this question is diminished if cryopumps for fusion application are configured to pump hydrogen isotopes and helium on different and separable surfaces within the pump.

4. Regenerable compound cryopump conceptual design

Preliminary concepts for the designs of tritium processing systems indicate a significant advantage in separating the bulk of the helium from the DT stream prior to delivery to the systems. This separation can be accomplished in a cryopump by pumping the DT and helium on different surfaces and subsequently warming the surfaces selectively by control of cryogen flow. A potential disadvantage to this approach is that, if there is an open path between the surfaces, the cryosorption surfaces must be warmed to a relatively high temperature to prevent sorbing of tritium during regeneration of the tritium pumping surface. Investigations of cryopump configurations were undertaken as part of a study by Leybold-Heraeus GmbH to define the NET vacuum pumping system [4].

This study defined the complete systems required for two alternative high vacuum pumping methods: cryogenic pumps, and turbomolecular pumps. A goal in the cryopump system design study was to minimize the number of individual pumps and cryogenic feed lines by designing single large pumps to meet pumping requirements through 16 pumped limiters. Design studies by the NET program aimed at the development of commensurately large isolation valves are in progress. Analyses of pumping requirements and pumping duct conductances indicate that the required pumping speed can be achieved by a 50 m^3/s cryopump on each pumping duct. NET requirements [4] specify the total tritium inventory in the cryopumps of 150 g maximum. This limit dictates periodic pump regeneration. To assure continuous system operation during cryopump regeneration, a second 50 m^3/s pump would be installed on each duct.

4.1. State-of-the-art of cryopumps

Prior to the design of the cryopump, a study was undertaken to determine the current state-of-the-art of cryopumps. The results of this study are presented in detail in ref. [4]. The study was performed by enumerating the latest advances in the parameters describing the characteristics of such pumps. Among the conclusions noted in the study were the following:

(a) Coconut charcoal appears to be the best sorbent for cryosorbing helium at 4.2 K. While zeolite will produce lower equilibrium pressures, charcoal will tolerate higher helium gas flow rates. Charcoal granules in the 12×30 mesh size (0.6–1.7 mm) produce the best results both in terms of pumping speed and capacity.

(b) The observed unit pumping speeds and capacities for helium were largely a function of the configuration of the thermal shield in front of the sorption panel. A pumping array with a charcoal-absorbing surface shielded by 2 chevrons will provide a unit pumping speed of approximately 4×10^{-3} $m^3/s/cm^2$ (4 $l/s/cm^2$). Tests with single liquid nitrogen chevron arrays have shown higher unit pumping speeds. A drop in pumping capacity for the single chevron system probably resulted from the fact that outer charcoal surface was slightly warmer because the charcoal viewed a warmer heat shield.

(c) Virtually no data are available on the sorption isotherms of tritium on charcoal.

(d) Data are needed on the long term effects of tritium and radiation on characoal or inorganic bonding methods.

4.2. Regeneration

The frequency of regeneration of the cryopumps is determined primarily by the maximum quantity of tritium permitted outside of the tritium containment area. During regeneration, all the previously pumped tritium should be dumped from the pump surfaces and exhausted to the tritium recovery stystem. Helium pumped on charcoal surfaces is effectively desorbed by raising the temperature to approximately 30 K [5]. This temperature is also expected to be adequate for removing DT from the condensation surfaces. It is, however, expected that the DT would be sorbed by the charcoal at this temperature. If, for example, the ultimate roughing pressure during regeneration were in the 10^{-2} Pa (10^{-4} Torr) range, it is estimated that the charcoal would require warming to approximately 100 K to avoid pumping of tritium. The need to warm to higher temperature increases the time required for warm-up and cool-down and increases the consumption of liquid helium. An alternative approach would be to prevent the tritium from reaching the sorption surface during regeneration.

4.3. Cryopump description

A cryopump has been designed incorporating a movable barrier that permits normal helium pumping during operation, and that moves to isolate the charcoal during regeneration. The pump shown in fig. 4 features a 1500 mm diameter inlet flange to coincide with the size valve being studied by the NET project team for future development. This pump has a cylindrical, charcoal-coated surface at 4.2 K, surrounded by a chevron heat shield also at 4.2 K, which in turn is surrounded by a solid liquid nitrogen-cooled heat shield with an entrance chevron.

A closed-end cylindrical barrier within the liquid nitrogen heat shield moves into place during regeneration to surround the charcoal surface, and isolates the surface from the remainder of the pump. The key

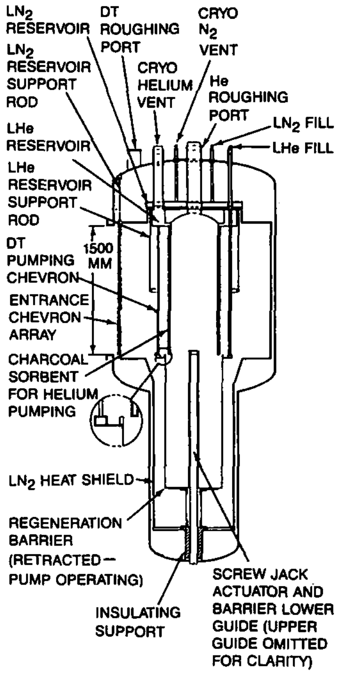

Fig. 4. Compound regenerable cryopump with movable barrier.

feature of this pump is that the liquid nitrogen-cooled heat shield is a totally sealed structure. As such, the only access of tritium to the pumping surfaces is by means of the entrance chevron. Thus tritium is prevented from bypassing the 4.2 K condensation surface and reaching the charcoal sorption surface. A baffle (shown in detail in fig. 4) incorporated in the barrier ensures that any tritium seeking to bypass the condensing chevron must make at least one contact with a 4.2 K surface. The cylindrical barrier is moved into place by a screw jack and pressed against a seat at the bottom of the liquid helium reservoir to assure minimum conductance in the "closed" position. Guides accurately position the barrier.

During regeneration, gases from the condensing surfaces are released into the pump volume; helium pumped by the charcoal is released into the barrier volume. By individually rough pumping these volumes, the helium is effectively separated from the DT stream prior to discharge to the tritium progressing system. The barrier also prevents impurities, such as tritiated hydrocarbons and water vapor that can cause degradation of the charcoal pumping ability, from reaching the charcoal during regeneration.

Preliminary pumping speed and conductance analyses indicate that the pump design shown will have a helium pumping speed in excess of the required 50 m^3/s. A Monte Carlo analysis shows the pump's helium speed to be approximately 95 m^3/s. A producibility analysis of the design indicates that the pump can be built and assembled.

4.4. Regeneration frequency

Sequential regeneration of the pumps was investigated as a means of maximizing time between regenerations and of smoothing the periodic dumping of regenerated gases and cryogenic helium. Analysis of a multiple station cryopump system with two pumps on each limiter, with one pump operating and one pump regenerating, indicates that the time (T) between regenerations of a given pump can be described by:

$$T = 2IG(Q(1.5G - 1))^{-1}$$

where:
I = maximum allowable tritium inventory, g.
Q = total tritium flow rate, g/h.
$G = N(N-1)^{-1}$.
N = total number of pumps.

For the NET pumping system with 32 pumps and a helium production rate of 2.4E20 atoms/s with a 5% He : DT ratio [6], the time between regenerations of an individual pump is 7.9 hr.

4.5. Cryogen distribution

The system required to distribute liquid helium to each cryopump and to vent it for regeneration is within the current state-of-the-art. Development is required only in the area of cryogenic bayonet connections capable of being operated by remote handling devices. The TFTR beamline system [7] is similar. Continuous operation of a cryogen supply system for pump regeneration was demonstrated with the CDC.

5. Conclusions

A charcoal-based system for application in fusion reactor cryopumps has been developed and a method for selecting charcoals for this application has been defined. A regenerable cryopump using the charcoal-based system has been conceptually designed to satisfy NET requirements. The design is based on near term state-of-the-art, and a demonstration model can be produced. The pump design avoids the uncertainty of co-pumping helium and hydrogen isotopes by incorporating a movable barrier. Development work is required to define charcoal characteristics in the presence of tritium, and to adapt current cryogenic bayonet designs for use with remote handling equipment.

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